Effect of SILAR Techniques on Photovoltaic Properties of PbS Quantum Dot Sensitized Solar Cells

W Yindeesuk[1], P Tusamalee[1], F Liu[2], T Toyoda[2] and Q Shen[2]

[1] Department of Physics, King Mongkut's Institute of Technology Ladkrabang, 1 Chalongkrung, Ladkrabang, Bangkok, Thailand.

[2] Department of Engineering Science, The University of Electro-communications, 1-5-1 Chofugaoka, Chofu, Tokyo 182-8585, Japan

E-mail: witoon.yi@kmitl.ac.th

Abstract. PbS quantum dots (QDs) have been received attention as a photosensitizer for quantum dot sensitized solar cells (QDSSCs) due to their narrow band gaps, quantum size effect, and multiple exciton generation (MEG). In this work, we prepared PbS QDSSCs by successive ionic layer adsorption and reaction method (SILAR) with PbS QDs deposited onto titanium dioxide (TiO_2), then the PbS QDs have been investigated the effect of SILAR cycles on photovoltaic properties. The power conversion efficiency (PCE) of the solar cells is dependent on SILAR cycles and an optimal SILAR cycle is two cycles. The PbS QDSSCs achieve the PCE of 1.79% with the photocurrent of 16.39 mA/cm^{-2}. Moreover, the optical properties were investigated by UV-visible absorption spectroscopy, which reveals a SILAR cycle affecting a band gap of PbS QDs.

1. Introduction

Quantum dot sensitized solar cell (QDSSC) is an area of intensive research due to the unique photoelectric properties of quantum dots (QDs), including size-dependent spectral tunability, excellent photostability, narrow emission spectrum, wide spectral response, high extinction coefficient, low fabrication costs, and multiple exciton generation. [1-3] With these advantages, the power conversion efficiency (PCE) of the QDSSCs can break the theoretical maximum from Shockley and Queisser limitation and potentially reach 44%. [4]

PbS is an important semiconductor material with a narrow direct band gap of 0.41 eV [5] and a large exciton Bohr radius, so it can change an energy band gap by controlling their QD size. In addition, PbS QD is one of the few types with absorption spectra down to NIR area, which can significantly improve the efficiency of solar cells. [6] When PbS QDs were prepared by successive ionic layer adsorption and reaction (SILAR) method, the particle size of the PbS QDs can be readily tuned by varying the number of SILAR cycles and hence their optical properties. [7]

In this research, we investigate the influence of SILAR cycles on the photovoltaic performance of the PbS QDSSCs. PbS QD sensitized TiO_2 photoanode was prepared by SILAR method, and then the PbS QDSSC was fabricated by sandwiching a liquid electrolyte containing polysulfide between the sensitized TiO_2 and Cu_2S which serves as the counter electrode. In experimental results, the solar cells with two PbS SILAR cycles exhibit the highest PCE of 1.79%.

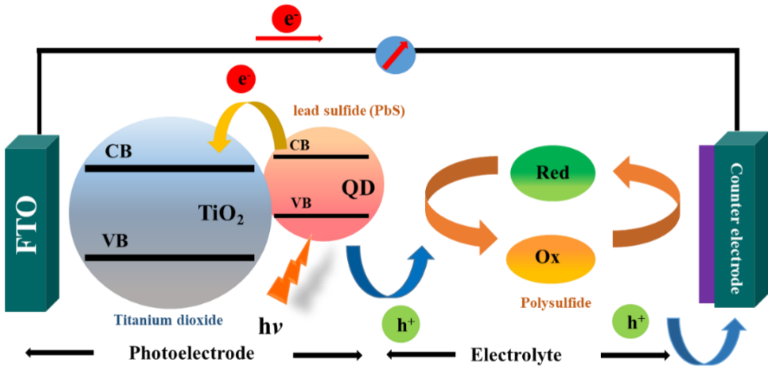

Figure 1. Schematic diagram of PbS QDSSC.

2. Experimental

Doctor blading method was used to prepare TiO_2 working electrodes on FTO (fluorine-doped tin oxide) glass. The films were sintered for 30 min at 200 ºC, 30 min at 350 ºC, and 30 min at 510 ºC to get a good electrical contact between nanoparticles. The thickness of TiO_2 was around 8-15 µm.

PbS QDs deposited on TiO_2 working electrodes were prepared by the SILAR technique. First, TiO_2 electrodes were dipped in Pb^{2+} precursor solution containing 0.02 M of lead acetate in methanol for 1 min. After that, the electrodes were rinsed with methanol. Then, the TiO_2 working electrodes were immersed in S^{2-} precursor solution containing 0.02 M of sodium sulfide in methanol/water (50/50, V/V) for 1 min, and then the electrodes were rinsed with methanol. These steps lead to one complete SILAR cycle. In this research, the PbS QDs with one, two, three, four, and five SILAR cycles were studied.

The polysulfide electrolytes were prepared using 1 M of Na_2S (sodium sulfide nonahydrate) and 1 M of S (Sulfur powder) in ultrapure water. The Cu_2S counter electrodes were prepared by immersing brass in HCl at 70 ºC for 10 min and dropping polysulfide solution for 10 min. [8]

The thickness of TiO_2 was measured by contact profilometer (Dektak, Ulvac). The cells were irradiated by AM 1.5G irradiation (100 $mWcm^{-2}$) with a Peccell solar simulator PEC-L10. The solar cells were assembled into sandwich type cells as shown in fig.1. The active area of the fabricated solar cells was 0.2376 cm^2.

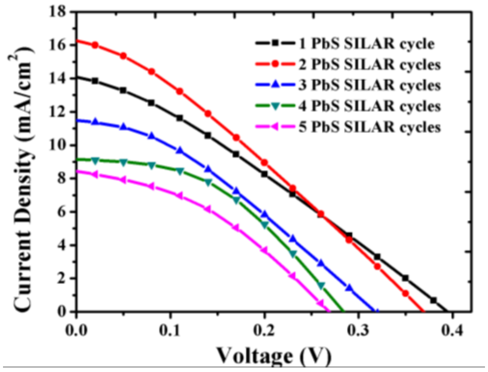

Figure 2. Photocurrent density-photovoltage (J-V) characteristics of PbS QD-sensitized solar cells with different PbS SILAR cycles.

3. Results and Discussion

PbS QDSSCs were prepared by the successive ionic layer adsorption and reaction (SILAR) method. Photovoltaic properties of the solar cells are evaluated with a current-voltage curve (J-V curve) which reflects solar cell performance by investigation between an applied voltage across the solar cell and the current response of the solar cell. The photocurrent density-voltage curves of PbS QDSSC were measured under illumination of 1 sun (AM 1.5, 100 mW/cm^2). The photocurrent density-voltage curves of PbS QDSSC with one, two, three, four, and five PbS SILAR cycles are shown in Fig.2. The photovoltaic parameters from J-V curve including, the open circuit voltage of the solar cell (V_{oc}), the short-circuit current of solar cells (J_{sc}), Fill factor (FF), and the power conversion efficiency of solar cells. The photovoltaics properties of the PbS QDSSCs are shown in Table 1. The highest efficiency of 1.79% was achieved in two PbS SILAR cycles.

The band gap of each QD can find from Tauc analysis. The estimated band gap from Tauc analysis was plotted in Fig.3(a). When SILAR cycles increased from one to five cycles, the bandgap of PbS QDs was decreased, corresponding to 2.52 eV, 2.08 eV, 1.68 eV, 1.56 eV, and 1.48 eV, respectively. The different band gap energy indicates the different sizes of QDs. The decreased band gap with increasing SILAR cycles shows that the increasing of QD size.

Table 1. Parameters of photovoltaics properties of PbS QDSSCs with different PbS SILAR cycles.

PbS SILAR cycles	J_{SC} (mA cm^{-2})	V_{OC}	*FF*	*PCE* (%)
1	14.20	0.39	0.29	1.65
2	16.39	0.37	0.29	1.79
3	11.54	0.32	0.33	1.23
4	9.17	0.28	0.44	1.14
5	8.52	0.27	0.38	0.86

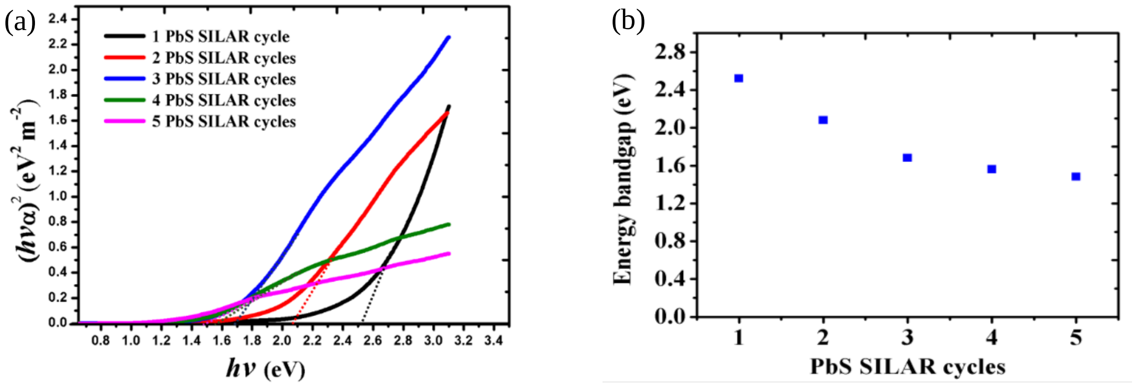

Figure 3. (a) Tauc plots of PbS quantum dot at different electrode SILAR cycles (b) Estimated band gap of PbS quantum dot at different PbS SILAR cycles.

The highest conversion efficiency of 1.79% was achieved with two PbS SILAR cycles. It is possible that the size of the quantum dot in solar cells which was prepared by two PbS SILAR cycles is suitable for electrolyte penetration into the QDs on TiO_2 photoelectrode. For this reason, the power conversion efficiency is highest. In case of one PbS SILAR cycle, it is possible that it has a small amount of PbS QDs. Consequently, the PCE of one PbS SILAR cycle is lower than PbS two SILAR cycles. In addition, when SILAR cycles increased, the power conversion efficiency decreased. The SILAR cycles affect the size of QDs. Therefore, when SILAR cycles increased, the size of PbS QD increased. Due to bigger QD size, the electrolyte may difficult to penetrate to QDs that are inside the TiO_2 electrode. Thus, the power conversion efficiency is lower than two PbS SILAR cycles.

4. Conclusions

PbS quantum dot sensitized solar cells are prepared by the successive ionic layer adsorption and reaction method. The effect of SILAR cycles on the photovoltaic performance of PbS quantum dot sensitized solar cells with one, two, three, four and five PbS SILAR cycles were investigated. In this study, the highest power conversion efficiency was achieved in two PbS SILAR cycles with power conversion efficiency of 1.79 %. The size of the quantum dot in solar cells prepared by two PbS SILAR cycles is the most suitable for electrolyte penetration into TiO_2 electrode. For this reason, the power conversion efficiency is highest. The estimated band gaps were 2.52 eV, 2.08 eV, 1.68 eV, 1.56 eV, and 1.48 eV with one, two, three, four, and five PbS SILAR cycles, respectively. We found that increasing PbS SILAR cycles lead to decreasing estimated optical bandgap due to quantum confinement effect.

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